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Preparation, Characterization and Fabrication of Eco-Friendly CZTS Thin Film Solar Cells by Vacuum Evaporation Method

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ABSTRACT

Non-toxic, eco friendly Cu_2ZnSnS_4 (CZTS) thin films, a suitable potential candidate for the absorber layers of solar cells, were successfully deposited on Mo coated glass substrate by thermal evaporation technique. CZTS thin film fabricates on the order of sequential vacuum evaporation of ZnS, Cu, and Sn. X-ray diffraction (XRD) studies revealed that polycrystalline films exhibiting kesterite structures with preferential orientations. The surface morphology, grain size and defects of CZTS films was determined from the scanning electron microscopy(SEM).The structural analysis showed increase with thickness, crystallanity and grain size increases whereas strain and dislocation density decreases. UV-Vis absorption studies revealed that CZTS thin film have an optical band gap from 1.4 – 1.56 eV, It was optimal for photovoltaic applications. A large absorption coefficient (10^4 cm^{-1}) and low resistivity achieved. The deposited film CZTS were annealed at 500°C, to avoid forming binary compounds. CZTS thin film based solar cell having fabrication consists of SLG/Mo/CZTS/CdS/i-ZnO/Al-ZnO/Ni/Al/MgF_2.The photovoltaic properties of CZTS thin film solar cells have been examined after formation.

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Keywords: Evaporation, X-ray diffraction, binary compounds, CZTS thin film.

1. INTRODUCTION

Development of green energy production has been depends on the peculiar properties of newer semiconducting materials. Thin film solar cells with various increasing efficiencies have been achieved by different kinds of techniques such as physical and chemical vapour deposition methods[1]. While a large part of this work has been focused on chalcopyrite and kesterite compounds[2], only a few semiconducting materials other than silicon like alloys can exist in the literature with good performance. Among them CIGS thin films have been synthesized and incorporated into high efficiency solar cells to improve the fill factor of the solar cells. Several other compounds like polymers[3] have also been reported although the Photovoltaic nature of these systems could limit, if any, their applications. It is thus necessary to look into other material with interesting optical, electrical and PV properties. CuInGaSe(Ga)Se thin film solar cells were prepared and reported[4].

Cu_2ZnSnS_4(CZTS) is one of the most promising absorber layer materials for low-cost thin film solar cells due to its semiconductor properties such as p-type conductivity[5], direct band gaps and high absorption coefficients ($>10^4$ cm^{-1}),as well as the abundant and nontoxic constituent elements. In the present work, we first utilized sequential vacuum evaporation technique to fabricate to form CZTS thin films. This method is more attractive than other ones in the photovoltaic applications because it is more suitable for large-scale production.

2. EXPERIMENTAL DETAILS

Cu_2ZnSnS_4 (CZTS) thin films were prepared by sequential vacuum evaporation method[6]. The films deposited onto glass substrates were first cleaned with detergent, water and then dipped in acetone. The metallic Cu, Sn and ZnS (purity 99.99%) were used as the evaporation sources. The chamber pressure was maintained at 10^{-5} to10^{-6} mbar during evaporation. The deposited film CZTS were annealed at 500°C, to avoid forming binary compounds. The films had a uniform thickness. The deposited CZTS thin films subjected to sulfurization in vacuum at 3hours[7]. To form p-n junction with the p-type CZTS, 50~100 nm n-type CdS thin film is deposited on the absorber layer by vacuum evaporation method. The surface of CZTS thin film is too rough to be fully covered by CdS thin film, leading to shortage between front contact and back contact. To prevent leakage intrinsic ZnO (i-ZnO) thin film is coated on CdS before 500~1000 nm transparent conducting oxide (TCO) thin film is deposited as the front contact layer of the cell.

The structural properties were determined by X-ray diffraction (XRD; Shimadzu) with Cu (Kα) radiation (λ = 1.5406 Å). Film morphology was analyzed by scanning electron microscope (SEM).The optical absorption and transmission spectra were obtained using a UV-vis spectro-

photometer within the wavelength range of 300nm to 1100 nm.

3. RESULTS AND DISCUSSION

3.1 XRD analysis

The phase composition of the sample was identified by XRD. As shown in Fig.1, the sample was of good crystalline nature and diffraction peaks of Cu_2ZnSnS_4 thin films can be conclusively indexed as tetragonal phase with the lattice constants of a=5.426Å and c=10.835 Å, which were consistent with the values in the standard card (JCPDS #26-0575). Sulfurization of the deposited CZTS thin films possesses a polycrystalline structure. Peaks attributed to (002), (101), (110), (103), (200), (105), (312) and (314) of CZTS appeared, which can be confirmed by the Powder Diffraction File (PDF) 026-0575. Moreover, the intensity of the (002) is the strongest, indicating that the sample corresponds to the kesterite structure which belongs to the tetragonal system. The XRD patterns of thin film samples indicated that not only the highly oriented and it has polycrystalline nature. The diffraction peaks of (002) and (101), indicates the presence of Copper Tin Sulfide[8].

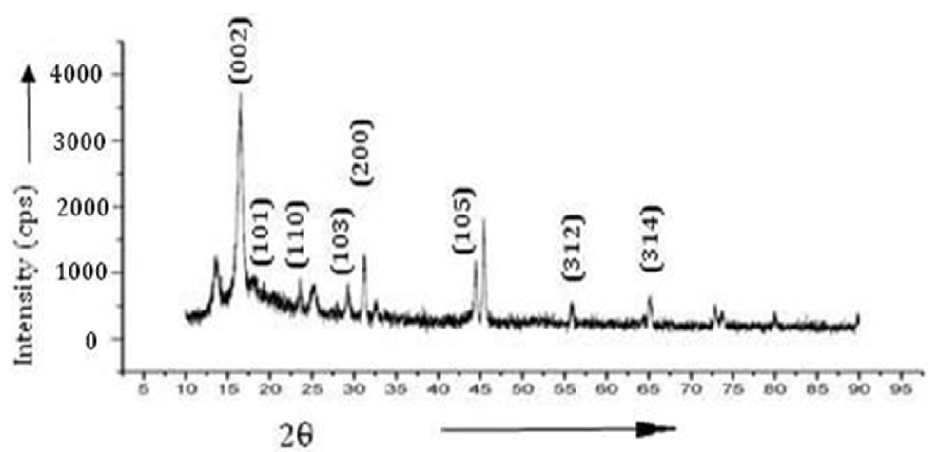

Fig.1 XRD pattern spectrum of the Cu_2ZnSnS_4 film

The crystallite size in Cu_2ZnSnS_4 thin film is evaluated from the intensity peaks of XRD by a Gaussian fit, using Debye-Scherrer formula[9].

$$D = \frac{0.98\,\lambda}{\beta Cos\,\theta} \qquad (1)$$

Where, β is the full width at half maximum, **λ** is the wavelength for X-ray used and θ is the Bragg’s angle.

3.2 Optical Properties

The optical absorbance spectrum was measured in the wavelength range of 400 – 700 nm using a Shimadzu spectrophotometer (Model -1800). Theory of optical absorption gives the relationship between the absorption coefficients α and the photon energy $h\nu$ for direct allowed transition as

$$(\alpha\, h\nu)^2 = A\,(h\nu - E_g) \qquad (2)$$

Where, A is a function of index of Fraction and hole/electron effective masses**.** The direct band gap is determined using this equation when linear portion of the $(\alpha\, h\nu)^2$ against $h\nu$ plot is extrapolated to intersect the energy axis at α =0. The value of optical band gap evaluated was ~1.47 eV[10]. The enhancement of band gap might be due quantum confinement arising from lowering of particle size.

3.3 Morphological analysis

SEM images, of p-type CZTS thin film of thicknesses ranging from 9 KÅ to 17 KÅ are shown in Fig. It has been found from Fig.2 that the films are fully covered, homogeneous, well adherent and free from crystal defects such as pin hole and cracks. The result shows that the film consists of compact structure grains with sub-micron size and low roughness, which is suitable for the absorber of thin film solar cells.

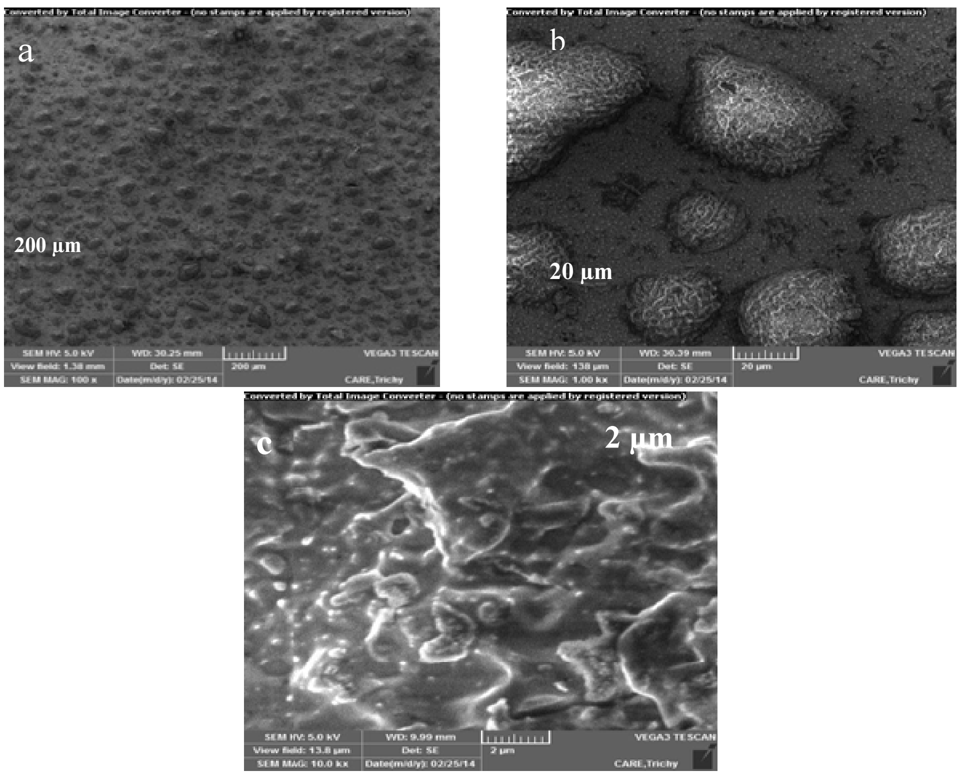

Figure 2: The SEM images of the CZTS thin film (a) 200 µm (b) 20 µm (c) 2 µm

4. CHARACTERIZATION OF THE CELL

The current-voltage (I-V) characteristic of the CZTS solar cell was measured with a source meter under one sun illumination intensity and the photovoltaic parameters are calculated. A conversion efficiency of 4.73% has been obtained for

the cell.

5. CONCLUSIONS

Eco-friendly p-CZTS/n-CdS thin film solar cells were successfully fabricated by sequential vacuum evaporation method. The sulfurization process which helps to increasing high crystalline phase of the deposited film. The deposited film CZTS were annealed at 500°C, to avoid forming binary compounds. When the ratios of the constituents are close to stoichiometric, the polyphase of kesterite type tetragonal structure was obtained. A high absorption coefficient as 10^4 cm^{-1} and an optical band-gap energy of the CZTS sample is about 1.47 eV can be achieved in this experiment, which is very close to the optimum value for a solar-cell absorber. The photocurrent characteristics of the optimized cell an efficiency of 4.73% are achieved. The experimental results showed that the current-voltage (I-V) characteristic properties of the cell have a strong dependence on the photovoltaic parameters of the thin films.

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